Experimental observation of a torus-doubling transition to chaos near the ferroelectric phase transition of a KH_2PO_4 crystal

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A ferroelectric KH_2PO_4 crystal is implemented in a simple series connection of an *RLC* circuit and the transition to chaos near the phase-transition temperature is investigated. The torus-doubling scenario to chaos, the theory of which was expounded by Kaneko for high-dimensional dynamical systems, has been found in the crystal. These experimental results suggest that observation of the nonlinear dynamical behavior in condensed matter can give much information about the correlation between the generation of nonlinearity and the order-parameter dynamics of the crystal near the phase-transition temperature. [S1063-651X(99)01311-2]

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I. INTRODUCTION

Since the time of Landau [1], the fate of a torus in phase-space flow has long been an active research interest in the field of nonlinear dynamics and chaos. According to him, a turbulent state can be reached by an infinite number of Hopf bifurcations, which introduces a new fundamental frequency into the dynamical system concerned [2]. Later this scenario was simplified by Ruelle and Takens [3]. They proposed that the system should undergo a transition from a fixed point into a quasiperiodic state with two-incommensurate frequencies (i.e., a flow on a two-torus) after two Hopf bifurcations. In this scenario chaos emerges when an infinitesimal perturbation or noise is introduced into the system. According to this scenario, two Hopf bifurcations from a fixed point are enough before the onset of chaos.

A quasiperiodicity associated with frequency locking in systems with two-incommensurate frequencies has been developed by Feigenbaum *et al.* [4]. This scenario shows a sequence of frequency locking events when the ratio of the two frequencies is rational, or quasiperiodicity when it is irrational before the onset of chaos. A map of a two-dimensional torus, i.e., a circle, is studied extensively by means of the so-called circle map to understand this scenario. Now quasiperiodic transition to chaos is a well-known scenario whereby a torus in low-dimensional dynamical systems ($d \leqslant 3$ in phase-space flow or $d \leqslant 2$ for a map) loses its stability and develops into chaos.

Another possibility for the fate of a torus, e.g., a cascade of period doublings of the torus, has been investigated numerically by Kaneko [5] in high-dimensional dynamical systems ($d \geqslant 3$ for maps or $d \geqslant 4$ for continuous flow). He combined two kinds of maps which belong to the two different universality classes, viz. period doubling and quasiperiodicity, associated with frequency locking, and found a new scenario, i.e., torus doubling, as a result of the interaction between the two scenarios. He also found that, unlike with the infinite number of period doubling from a fixed point, the torus undergoes a finite number of doublings before the onset of chaos.

This scenario has been reported in many numerical experiments by Franceschini [6] on seven-mode Navier-Stokes equations on a three-dimensional map by Arnéodo *et al.* [7], and on a quintic complex Ginzburg-Landau equation by Kim *et al.* [8]. Though the existence of a torus-doubling scenario has been pointed out in theory [5,9], it is believed to be difficult to observe in experiments. But exceptions can be found in the recent experiment of Rayleigh-Bénard convection [10], in the convection of molten gallium [11], and in a chemical reaction [12].

All of those systems reporting torus-doubling phenomena are infinite-dimensional dynamical systems. Here we report on the torus-doubling phenomena in condensed-matter physics near the phase transition of ferroelectric KH_2PO_4 (KDP) crystal. A few works [13,14] can be found on the torus-doubling scenario in the areas of condensed-matter physics. In a KDP crystal near the phase transition, fluctuations in the order parameter P are described by a highly nonlinear Duffing oscillator [15], a representative example of a dynamical system exhibiting period doubling. This crystal also displays piezoelectricity near the phase-transition temperature between the order-parameter fluctuation P and the elastic shear strain X. A real physical model of the KDP crystal near the phase transition utilizing a simple *RLC* circuit is described in detail in Sec. III. Thus the nonlinear dynamical behavior of the KDP crystal near the phase transition is described by two-anharmonic oscillators coupled by an electromechanical coupling, each of which is a representative example of dynamical systems showing period doubling and quasiperiodicity. Regarding these facts, KDP crystal is a good example of a nonlinear dynamical system in condensed matter for studying the interaction between period doubling and quasiperiodicity.

This paper is organized as follows. In Sec. II the experimental setup is described. In Sec. III the physical model of the system is given and the experimental observations of torus-doubling phenomena in the first return map, real-time waveform, and power spectrum are presented in Sec. IV. Section V summarizes our main conclusions.

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II. EXPERIMENTAL SETUP

For our experiment, KDP samples were prepared in two stages. First the crystal was cut perpendicular to the *c* axis to get a thin slab and then cut again along the 45° diagonal in the *ab* plane to couple the piezoelectric excitation to the *c*-axis polarization of the crystal. Typical dimensions of the sample were $7.1\times 3.5\times 0.5\ \mathrm{mm}^3$. To make electrodes on the sample, it was evaporated with gold and then painted with silver paste. With the sample geometry prepared in this way, a shear strain X_6 is coupled piezoelectrically to the polarization P along the c axis so it simply dilates along the length. The KDP samples prepared were used as nonlinear capacitors in a simple RLC resonator with $L=10$ mH and $R=390\ \Omega$. We used cryogenic equipment (a closed-cycle helium refrigerator and a cryostat) provided as a package by the JANIS Research Co., Inc. We also used a Lakeshore 330 autotuning temperature controller whose control stability is given by ± 25 mK at 300 K with a silicone diode sensor. With the cryostat set at 3–5 m Torr, the temperature of the KDP sample was controlled as finely as ± 0.01 K around T_c. As a signal generator, we used an HP3325B function generator whose resolution is given by 1 μHz under 100 kHz from 20 to 30 °C. To characterize the sample, we performed the dielectric measurements first. An HP4275 multifrequency spectrum analyzer was used at $f=10$ kHz and $T=150$–50 K in steps of 1 K. Then we did the same experiments again in steps of 0.02 K near T_c to determine exactly the phase-transition temperature of the KDP crystal. T_c was found to be 121.69 K and the voltage signal across the resistor was measured as a dynamical variable.

A Poincaré map, a stroboscopic motion of the trajectory on a section plane in phase space, is a common way of displaying the dynamics of quasiperiodicity and period doubling of the torus. On the Poincaré section, a two-frequency quasiperiodic motion in the continuous flow forms a circular ring on the Poincaré section plane while a period-doubled torus is represented by a breakup of the circular ring into two rings. Experimentally, a Poincaré section or return map is generated by a series connection of a sample and hold circuit, which holds the maximum of the signal with one period delayed in succession. Additionally, a pulse generator is used to trigger the signal at the peak position. It consists of a differentiator and a zero-crossing detector. These pulses are used to strobe the Z axis of the oscilloscope to display clearly the Poincaré section of the phase plots on a cathode-ray tube. An HP54600A image storage digital oscilloscope is used to capture the image generated in this way. To analyze the temporal behavior of the dynamics, an image of the real-time waveform is also captured on the cathode-ray tube and regulary spaced time series data as long as 2000 points have been recorded for later processing of the temporal Fourier spectra.

III. PHYSICAL MODEL OF THE KDP CRYSTAL NEAR PHASE TRANSITION IN AN *RLC* CIRCUIT

Now let us suppose that a KDP crystal is implemented in a simpe RLC series resonance circuit as a nonlinear capacitor and it is driven by a sinusoidal driving voltage, $V(t)=V_o\sin(2\pi ft)$, provided by a signal generator HP3325B. By an application of Kirchhoff's law, the equation of motion for the order parameter P can be derived and it is given by the Duffing's nonlinear oscillator equation [15]. A lot of numerical studies report that it is a representative example of a dynamical system which shows a finite number of period doublings [16]. For the KDP sample used in our experiment, shear strain X_6 is coupled electromechanically to the polarization P along the c axis of the crystal. Regarding the simplest bilinear coupling, ηPX with η=coupling strength, the following equation of motion is proposed [15],

$$\begin{gathered}\ddot{P}+a\dot{P}-P+P^3+\eta X=\nu\sin(\Omega\tau),\\ \ddot{X}+b\dot{X}-X+X^3+\eta P=0,\end{gathered}\tag{1}$$

where τ is the dimensionless time, a and b are the damping constants related to the polarization fluctuation and shear strain, Ω is the dimensionless frequency, and the differentiation is with respect to τ. According to Eq. (1), the dynamical behavior of a KDP crystal near the phase transition is described by two-anharmonic oscillators, a polarization and a shear-strain oscillator, coupled by an electromechanical coupling, each of which is a representative example of period doubling and quasiperiodicity. When it is written as a set of first-order equations,

$$\begin{gathered}\frac{dP}{d\tau}=Q,\quad \frac{dQ}{d\tau}=-aQ+P-P^3-\eta X+\nu\sin\phi,\\ \frac{dX}{d\tau}=Y,\quad \frac{dY}{d\tau}=bY+X-X^3-\eta P,\\ \frac{d\phi}{d\tau}=\Omega,\end{gathered}\tag{2}$$

to analyze the dynamical behaviors, it becomes a five-dimensional dynamical system in continuous phase flow and thus satisfies the necessary condition for the torus-doubling transition to chaos.

Four-dimensional Poincaré maps of the type

$$\begin{aligned}P_{n+1}&=f_1(P_n,Q_n,X_n,Y_n),\\ Q_{n+1}&=f_2(P_n,Q_n,X_n,Y_n),\\ X_{n+1}&=f_3(P_n,Q_n,X_n,Y_n),\\ Y_{n+1}&=f_4(P_n,Q_n,X_n,Y_n),\end{aligned}\tag{3}$$

can be constructed by a Poincaré section technique and will be given by a combination of maps which belong to the two different universality classes: period doubling and quasiperiodicity associated with frequency locking. It can be found from Eqs. (2) and (3) that a KDP crystal near the phase transition serves as a good example of a nonlinear dynamical system in condensed matter to study the interaction between the two universality classes: period doubling and quasiperiodicity. The mappings given by Eq. (3) can sometimes be simplified into low-dimensional maps showing period doubling or quasiperiodicity when one of the variables can be written as a function of the other three variables. It should also be pointed out that even though an exact analytic form

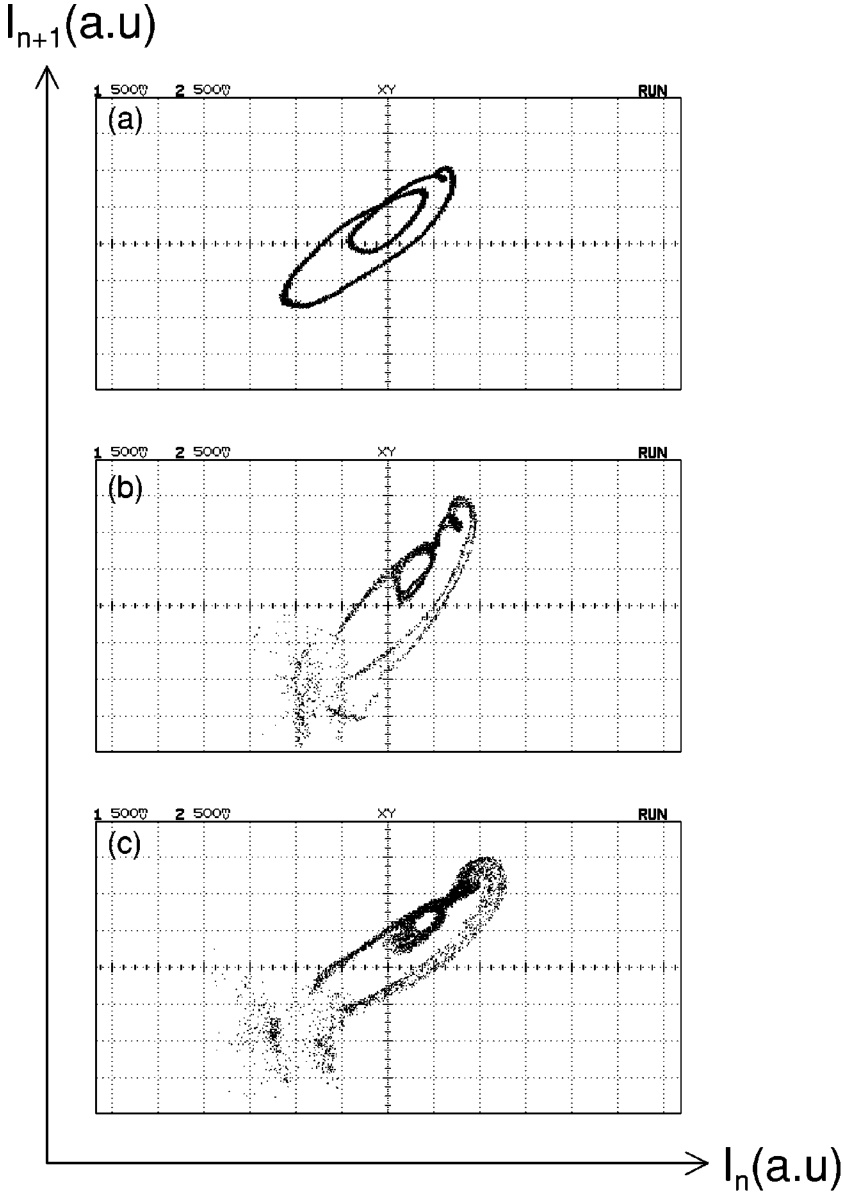

FIG. 1. I_{n+1} vs I_n. I_n is a local maximum of the current oscillation across the resistor. (a) Torus generated by the Hopf bifurcation at $f = 64.395$ kHz. (b) Period-doubled torus at $f = 64.431$ kHz. (c) Chaos at $f = 64.453$ kHz.

of the equation is not available, maps obtained in experiments can be useful in illustrating the bifurcation nature of the dynamical system concerned. In the experiment, we have observed period doubling of a fixed point, quasiperiodicity, and period doubling of a torus, as will be shown below.

IV. OBSERVATION OF TORUS-DOUBLING PHENOMENA

A. Poincaré section

In our experiments, period doubling of the torus has been obseved as the frequency of the external stimulus is increased. The current oscillation of the periodic state at the driving frequency, f_1, in the KDP crystal undergoes a Hopf bifurcation into a quasiperiodic state with two-incommensurate frequencies, f_1 and f_2. The second self-oscillation frequency, f_2, in the crystal is generated by the piezoelectricity provided by an electromechanical coupling between the order parameter P and the shear strain X of the crystal.

For quasiperiodic motion with two-incommensurate frequencies, f_1 and f_2, the first return map looks like a closed loop since the trajectory fills up the circle completely in an ergodic way. So a closed circle in the first return map as shown in Fig. 1(a) at $T = T_c + 0.1$ K indicates that the motion is indeed quasiperiodic with two-incommensurate frequencies inherent in the system.

After the Hopf bifurction, the torus undergoes a period doubling in the beat (or modulation) frequency $\Delta f = f_2 - f_1$ and this is presented in the first return map [Fig. 1(b)]. The circular ring breaks into two rings, each of which is visited alternately by the trajectory. After a finite number of period doubling events, chaos quickly appears and the two rings in the first return map merge, forming a circular band as is

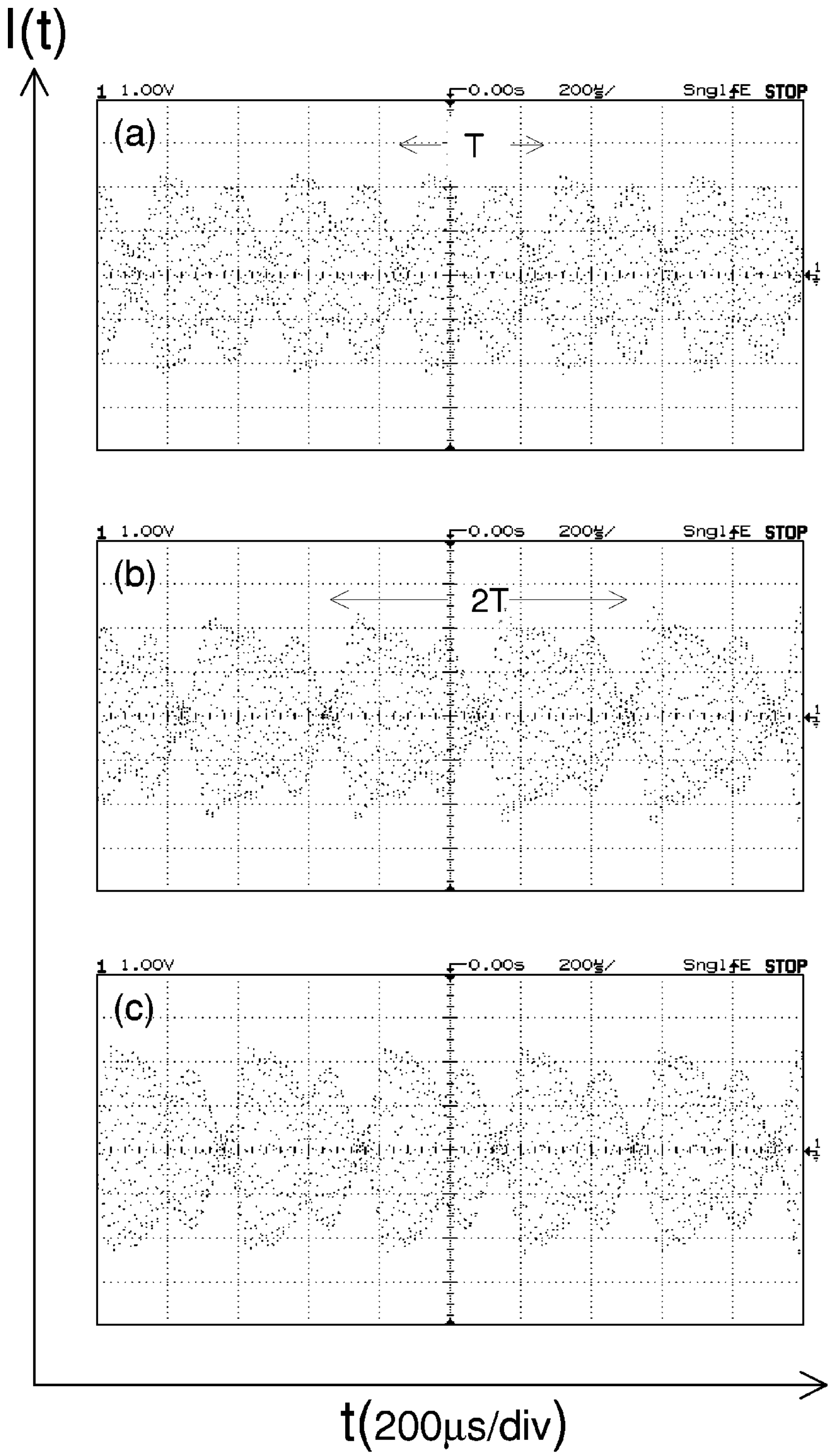

FIG. 2. Corresponding real-time waveform of the current oscillation. (a) Quasiperiodic oscillation (torus) with external driving frequency f_1 and piezoelectric self-oscillation frequency f_2 generated by an electromechanical coupling. The beat frequency in the modulation amplitude is given by $\Delta f = f_2 - f_1$. (b) Current oscillation of the period-doubled torus. This state corresponds to the quasiperiodic oscillation with two-independent frequencies of f_1 and $f_3 = f_1 + \Delta f/2$. (c) Current oscillation of the chaotic state.

shown in Fig. 1(c). The whole series of the evolution of the torus, as is shown in Fig. 1 from (a) to (c), is believed to be strong evidence for a transition to chaos via the period-doubling scenario of a torus in a high-dimensional dynamical system.

A real-time waveform of the modulated signal, $V_R(t)$, is displayed in Fig. 2. As will be seen from the figure, the envelope of the waveform varies slowly with time at the beat frequency $\Delta f = f_2 - f_1$ in a sinusoidal way. Therefore if the response signal $V_R(t)$ is initially different from zero, as time increases, the envelope of the amplitude of the signal $V_R(t)$ decreases slowly with time and nearly becomes zero. But, when $V_R(t) \geqslant 12.0$ or $V_R(t) \leqslant 0.5$ V, sample and hold amplifiers cannot hold the input signal exactly and this is the reason why the trajectories are scattered like noise in the third quadrant of the first return map.

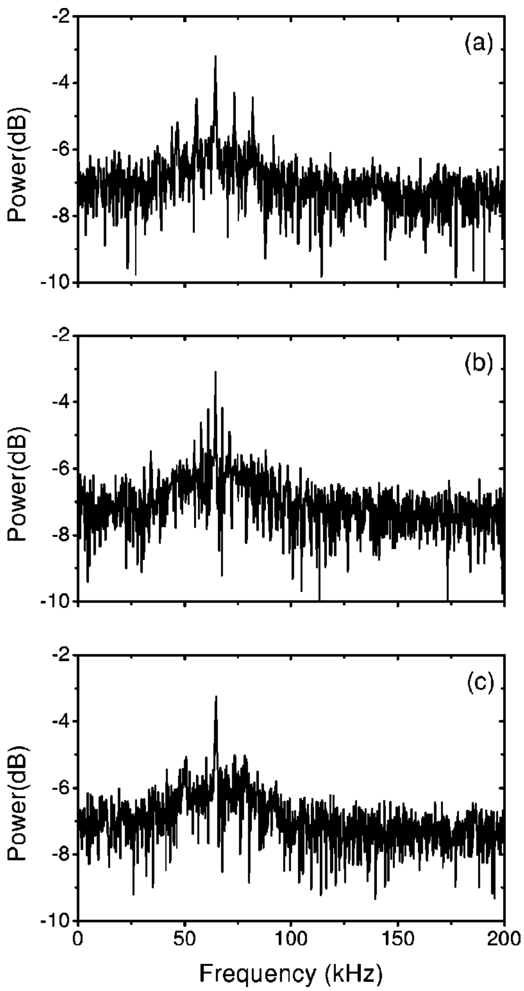

FIG. 3. Corresponding power spectra of the time series in Fig. 2. (a) Quasiperiodic state of two-incommensurate frequencies with f_1 and f_2. (b) State of the period-doubled torus: quasiperiodic state with f_1 and $f_3 = f_1 + \Delta f/2$. (c) Chaotic state.

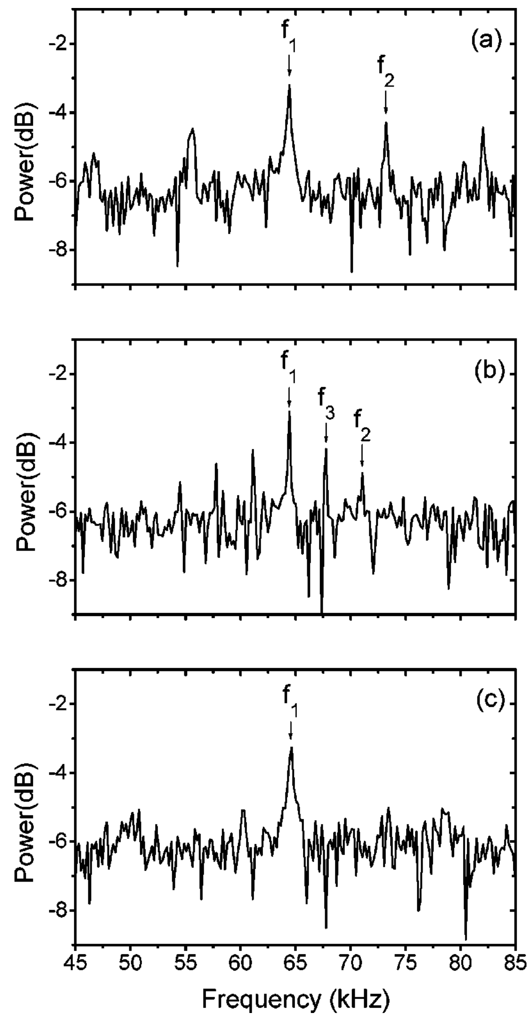

FIG. 4. Enlarged power spectra of Fig. 3. (a) Quasiperiodic state with f_1 and f_2. (b) State of the period-doubled torus: quasiperiodic state with f_1 and $f_3 = f_1 + \Delta f/2$. (c) Chaotic state.

B. Real-time waveform

In Fig. 2, the corresponding real-time waveform of the current oscillation is given. As the frequency starts to increase, the current in the circuit oscillates at the frequency, say f_1, of the external driving voltage signal. As the frequency is increased further, the current undergoes a Hopf bifurcation caused by the piezoelectric coupling between the polarization and the strain of the ferroelectric KDP crystal. This gives the second self-oscillation frequency, f_2, in the system and, as a result, the amplitude is modulated [Fig. 2(a)] with the frequency given by $\Delta f = f_2 - f_1$.

The period of the beat in the amplitude modulation is given by $T = 1/\Delta f$. In the time series shown in Fig. 2(b), when the torus doubling happens, it is also doubled and this phenomenon can be seen more clearly in the corresponding power spectrum. This is shown in Figs. 3 and 4 in detail and, in the power spectrum, torus doubling means the generation of a new subharmonic frequency at $f_3 = f_1 + \Delta f/2$, midway between the two fundamental frequencies, f_1 and f_2. So, in the torus-doubled state, all the other frequencies can be expressed as a linear combination of two independent frequencies, f_1 and f_3, instead of f_1 and $f_2 = f_1 + \Delta f$. Futher increase of the frequency leads to the onset of chaos in the modulation of the beat [Fig. 2(c)].

C. Power spectrum

The corresponding power spectrum and its enlargement are given in Figs. 3 and 4, respectively. When the crystal is driven away from the piezoelectric resonance, the current response of the crystal oscillates with the external driving frequency f_1. As f_1 is increased further towards the piezoelectric resonance frequency, the crystal undergoes a Hopf bifurcation and an additional self-oscillation frequency, f_2, appears spontaneously in the system. Since the temperature of the KDP crystal is fixed, the piezoelectric oscillation frequency, f_2, is essentially frequency-dependent on f_1 of the signal generator. Due to the modulation between the two frequencies via an electromechanical coupling, the voltage oscillation across the resistor, $V_R(t)$, gives a beat in the amplitude modulation with frequency $\Delta f = f_2 - f_1$, shown in the corresponding power spectrum in Fig. 4(a). Since f_1 and f_2 are two-independent frequencies, f_1 and Δf are also independent. So all the other frequencies present in the system can be expressed as a linear combination of them.

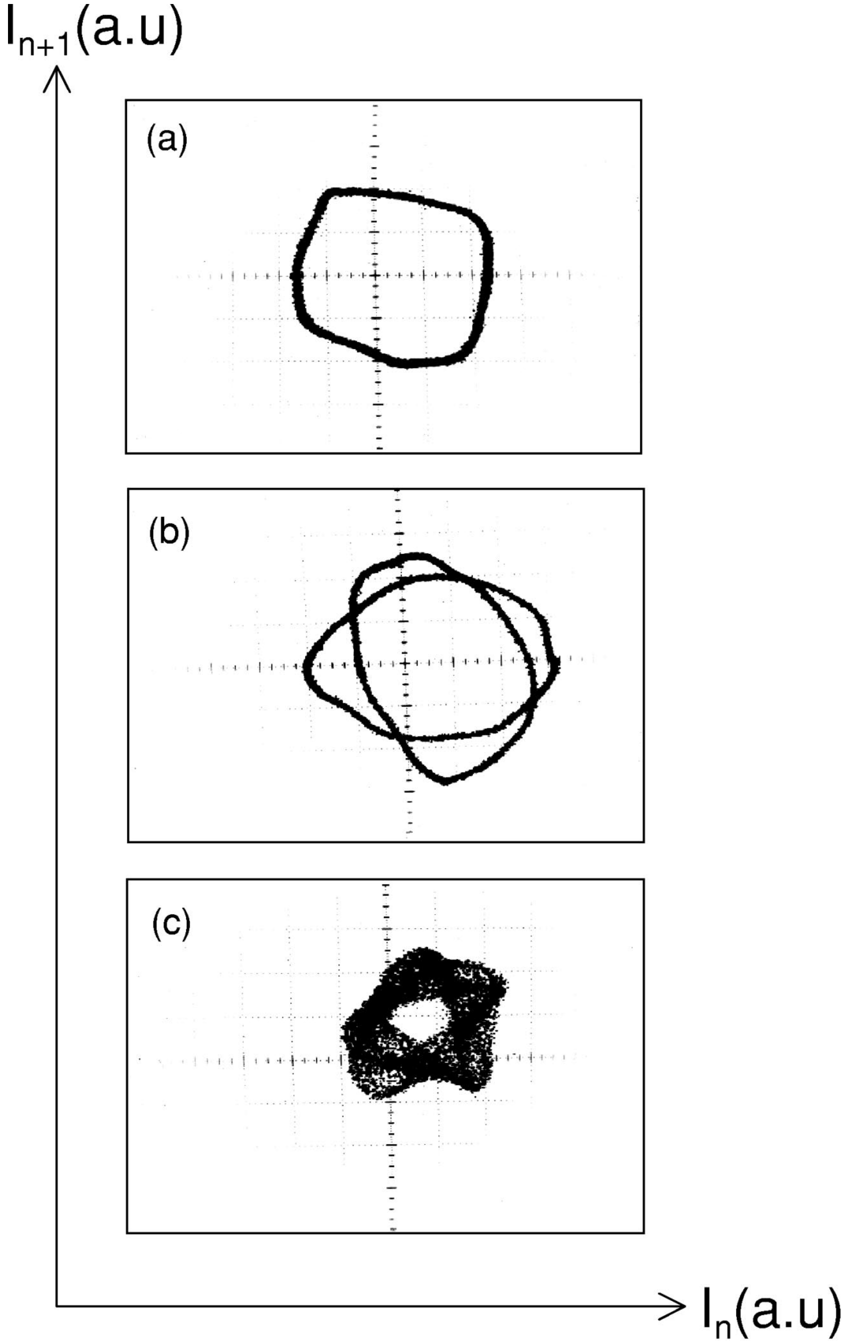

FIG. 5. Separated case of torus doubling: (a) torus at $f = 62.092$ kHz, (b) period-doubled torus at $f = 61.074$ kHz, and (c) chaos at $f = 60.404$ kHz.

When the period of the amplitude modulation is doubled, the corresponding beat frequency is halved to $\Delta f/2$ and a new peak in the power spectrum appears midway between the two frequencies, f_1 and f_2. For this period-doubled state of the torus in the system, the new subharmonic frequency at $f_3 = f_1 + \Delta f/2$ becomes their new beat frequency. In this way, whenever there is a cascade of period doublings of the torus, the beat frequency is halved and more subharmonic frequencies are generated in the power spectrum. So the appearance of new subharmonic frequencies at $f_3 = f_1 + \Delta f/2$, midway between the two fundamental frequencies f_1 and f_2 of the system, is a strong indication of the onset of period doubling of the torus.

Doubling of the torus terminates after a finite number and then chaos appears. All the sharp peaks in the power spectrum are smoothed out and the power spectrum becomes nearly flat [Fig. 4(c)], a characteristic feature of chaotic motion in the power spectrum.

D. Temperature dependence of the phenomena

In our investigation, torus-doubling phenomena have been observed within ± 1 K of T_c. In the paraelectric neighborhood, when $T \leqslant T_c + 1.0$ K, we frequently observe period doubling, quasiperiodicity associated with frequency locking, and torus doubling. But, when $T \geqslant T_c + 1.0$ K, these nonlinear dynamical responses are hardly observed. On the

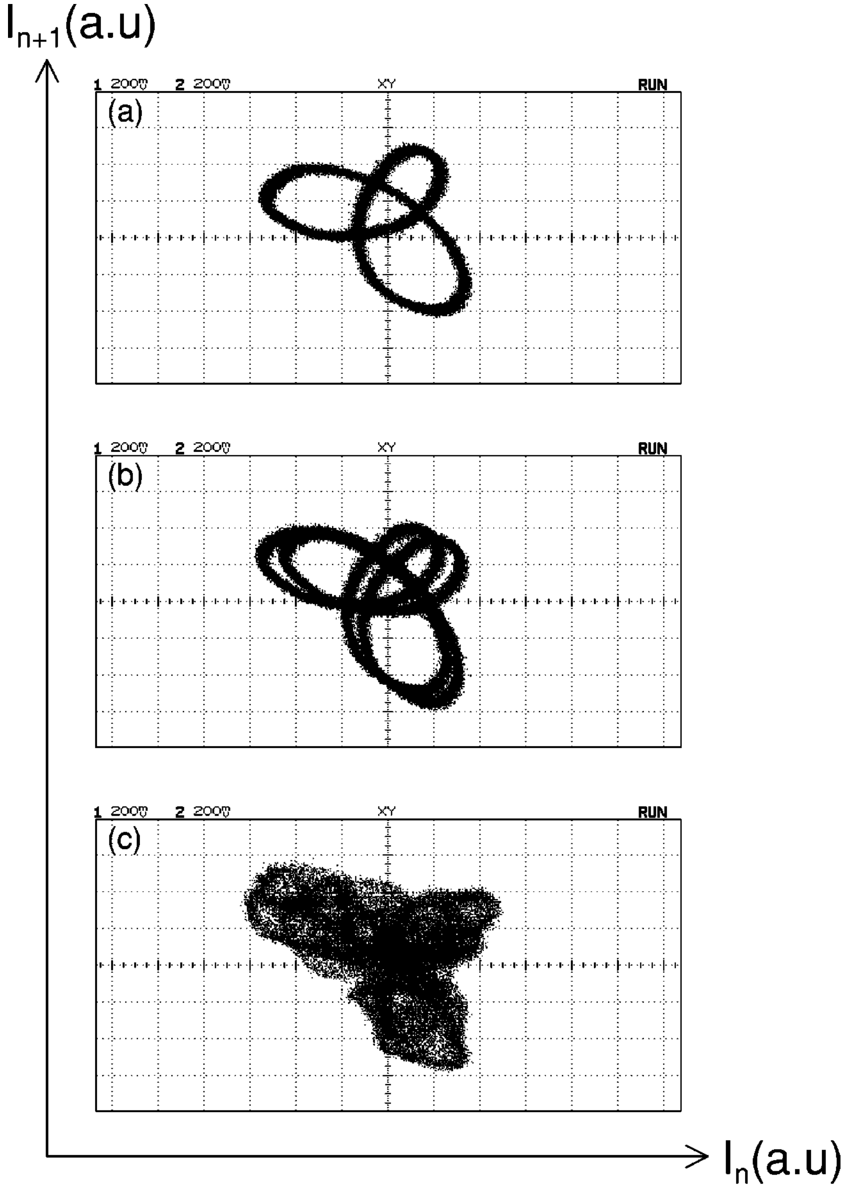

FIG. 6. Folded and separated case of torus doubling: (a) torus at $f=56.522$ kHz, (b) period-doubled torus at $f=56.538$ kHz, and (c) chaos at $f=56.547$ kHz.

other hand, in the ferroelectric neighborhood, these nonlinear phenomena can be observed down to $T \geqslant T_c - 5.0$ K, depending on the sample. We guess that this is related to the large domain wall motion [17] of the sample and, as a result, a large polarization fluctuation in the sample down to the domain wall freezing temperature.

E. Other types of torus-doubling phenomena in Poincaré section

Kaneko [5] noted that two types of torus doubling are possible in his numerical study. One is the case in which the cross section of a torus is separated and the other is the case in which it is still connected but folded. In our experiment we have found both types of torus doubling. In some cases the torus is separated [Fig. 5(a)] and in other cases it is folded and separated simultaneously [Fig. 6(b)] depending on the KDP samples prepared.

F. Further doublings and characterization of the onset of chaos

In the torus-doubling transition into chaos, it is not clear whether the doubling cascade can continue infinitely in genetics. In theory, it has been found that the doubling cascade stops after a finite number of times and then chaos appears with the fractalization of the torus at the onset of chaos [5]. In all examples of the torus-doubling transition into chaos observed so far, only a finite number of doublings have been reported. In our experiment, maximally two times of the torus doublings have been observed.

V. SUMMARY

In summary, we have observed an unusual transition to chaos near the ferroelectric phase transition of KDP. The current oscillation through the KDP crystal undergoes a transition from a fixed point to a quasiperiodic motion with two-incommensurate frequencies (i.e., a flow on a two-torus) through a Hopf bifurcation, to a quasiperiodic motion on a period-doubled torus through a cascade of torus doublings, and then to chaos, which has been predicted theoretically by Kaneko [5]. Significantly, we have found this phenomenon near the ferroelectric phase transition of a KH_2PO_4 crystal in condensed matter. Torus-doubling phenomena in a KDP crystal are associated with the order-parameter fluctuation, modeled by Duffing's nonlinear oscillator, and the piezoelectricity of the crystal between the soft-mode polarization P and the shear strain X of the KDP crystal. Observation of a finite number of period doublings of the torus in the crystal suggests that polarization fluctuation as well as an electromechanical coupling due to the large atomic displacement of the crystal near the phase transition plays a crucial role in the nonlinear dynamical behavior of the crystal. These results show that study on the nonlinear dynamical behaviors of the condensed matter gives much information on the correlation between the generation of nonlinearity and the order-parameter dynamics near the phase-transition temperature. Now it is our conjecture that a torus-doubling transition to chaos can be observed frequently in most ferroelectric materials because they have a strong electromechanical coupling between the piezoelectric oscillation and the large order-parameter fluctuation near the phase transition.

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